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## Verification with gamma-ray and fast neutron detection

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### Abstract

We present the recent results of our development effort with liquid scintillators at Lawrence Livermore National Laboratory. Historically we developed many assay analysis techniques specifically for relatively low efficiency (a few percent) neutron measurement systems using thermal neutron detectors, mainly helium-3, taking advantage of the high thermal neutron interaction cross-sections. Recently, however, we have been investigating fast neutron detection with liquid scintillators and inorganic crystals. We have discovered considerable advantages with fast neutron detection because the inherent nanosecond production time-scales of fission and neutron-induced fission are preserved instead of being lost in neutron thermalization required for thermal neutron detectors.

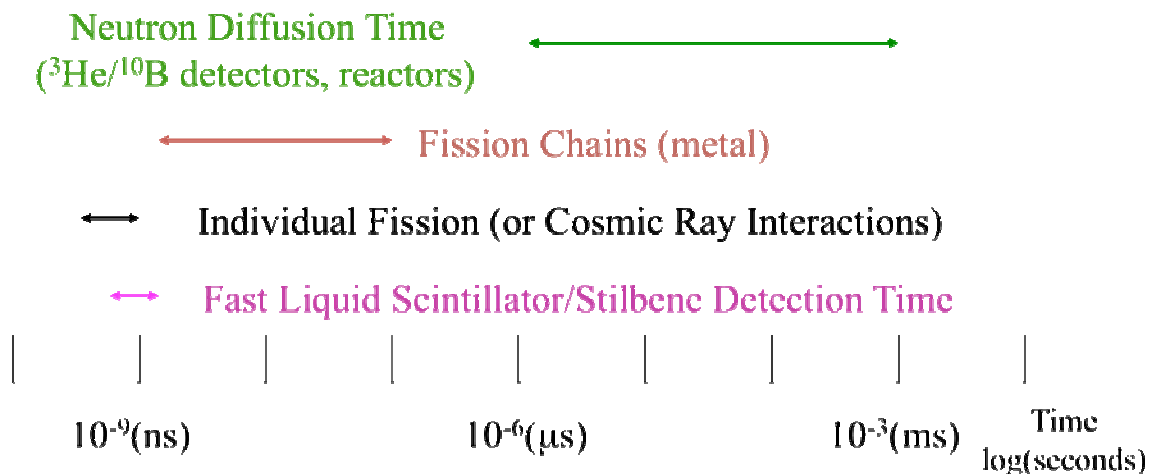
### Introduction

Weapons verification requires the ability to confirm attributes like the type of material (elements, isotopes), the amount of material (mass), and its configuration (shapes). This means the ability to distinguish Pu from non-Pu, Pu from Pu oxide, weapons-grade Pu from non-weapons-grade, balls from shells, and multiple weapons from single weapons. Existing detection systems can make some of these distinctions. For instance, high-resolution gamma spectrometers can detect isotopic information from gammas that are not too highly shielded or self-shielded. Thermal neutron detectors, like helium-3 tubes encased in polyethylene, can obtain neutron flux and, when the intrinsic and geometric efficiency is high, they can also obtain multiplication. Time-evolution of correlated thermal neutrons can also indicate the presence of moderating substances. Further, gamma flux is an indicator of surface area, and neutron multiplication is an indicator of volume, and these two parameters put together give an idea of geometry and density. There are also a few methods for obtaining imaging passively with gammas and neutrons. We report on capabilities of a liquid scintillator detector system that provide complementary and additional capabilities for weapons verification.

### Characterization Supported by Theory

We have chosen to understand what we expect to see. We create this understanding using a detailed theory of fission chains. Prior to this project, we developed a reconstruction theory for thermal neutron counting, based on a statistical theory of fission chains. This theory assumed the fission chain was instantaneous compared

to diffusion time scales. The theory enabled us to do absolute assay of unknown simple objects. In some cases, though, the reconstruction could be degenerate. Sometimes it was only possible to determine a relation between detection efficiency and multiplication. Sometimes it was difficult to determine if a fission chain was initiated by spontaneous fission, by (alpha, n) reactions, or by cosmic rays. (The spontaneous fission rate is proportional to the mass of the dominant spontaneous fission isotope.) For HEU problems cosmic rays (only approximately modeled, but included in the reconstruction theory) could make quantitative assay difficult. For some problems the neutron signal was sufficiently absorbed that correlation information was lost. In some cases the assumptions of the underlying thermal neutron theory were inadequate for the object. Real fissioning items emit time-evolving neutron and gamma-ray signals with time and energy correlations that have not yet been fully exploited.



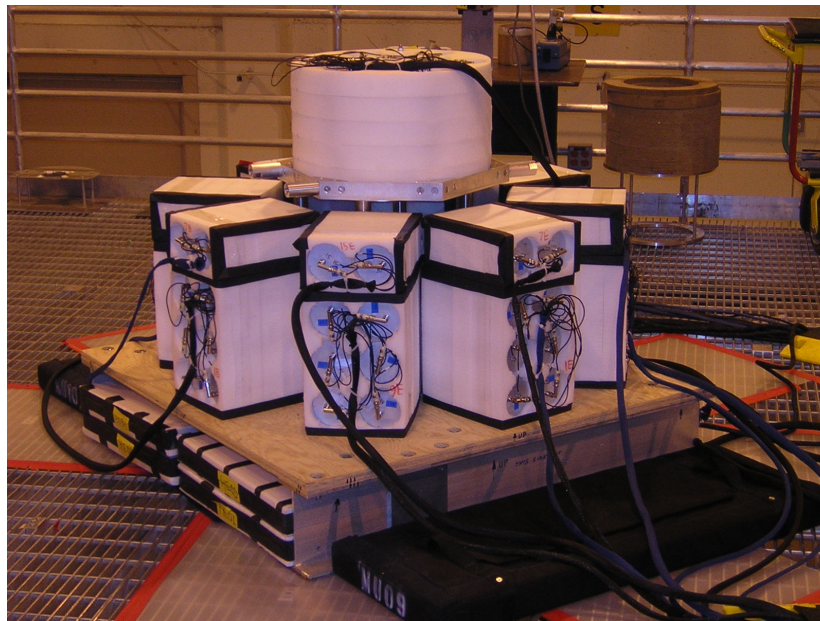
**Figure 1: Timescales of different processes and two varieties of neutron detectors.**

This past year we expanded our theoretical understanding to include the time evolution of fission chains because we have begun to work with liquid scintillators with nanosecond time-tagging electronics. Combined with thermal neutron and time-tagged HPGe spectroscopy, many additional fission chain correlations can be measured. Liquid scintillators (LS) based on xylene are sensitive to gammas, fast neutrons, and minimum ionizing particles (MIPs) associated with cosmic rays. The time-taggers allow us to associated individual particles with individual events (Figure 1). The new theory can possibly enable new assay methods beyond traditional methods based on various forms of statistical counting distributions, although there is now more information in these distributions. For example, the fast fission chain time constant and multiplication of SNM metal can be determined independently of the system multiplication, using the simplest form of the theory that generalizes Feynman moments analysis to the shake (1 shake = 10 nanoseconds) time scale. As another example, counting gamma-rays from the fast fission chain or from (n, n' gamma) are on such a fast time scale that the high gamma-ray background becomes negligible. With an appropriate theoretical framework these contributions can also be used for assay. Fast counting can isolate

individual spontaneous fission events, even for high source rate. The ability to count both the fast neutrons and gamma-rays from such events may help distinguish (alpha, n) sources from spontaneous fission sources. (An imaging method made possible by this capability will be discussed below.) For thermal neutron counting even multiple spontaneous fission events overlap on a diffusion time scale. With fast counting individual fission chains in Pu can be isolated, and easily distinguished from strong spontaneous fission sources like Cf.

### **Liquid Scintillator Detector Array**

Scintillator detector manufacturers offer a few liquid scintillators with different properties. We have been working with Eljen Technology's EJ-301 (also known as NE-213 and BC-501A) assembled in cylindrical modules that are 3-inches high and 4-inches in diameter. In brief, these scintillators provide fast response with one nanosecond arrival time resolution and can discriminate gammas from fast neutrons. They also have energy resolution around 20% and approximately 50% detection efficiency for neutrons of a few MeV [1]. These small modules give only a Compton spectrum for detected gammas. They also give low-resolution neutron spectroscopy by conversion of gamma energy to neutron energy by means of a "quench" function. Much like gamma Compton spectra, the computed neutron energy represents a lower bound on the incoming neutrons' original energy, which is often much higher (see for example [1]).



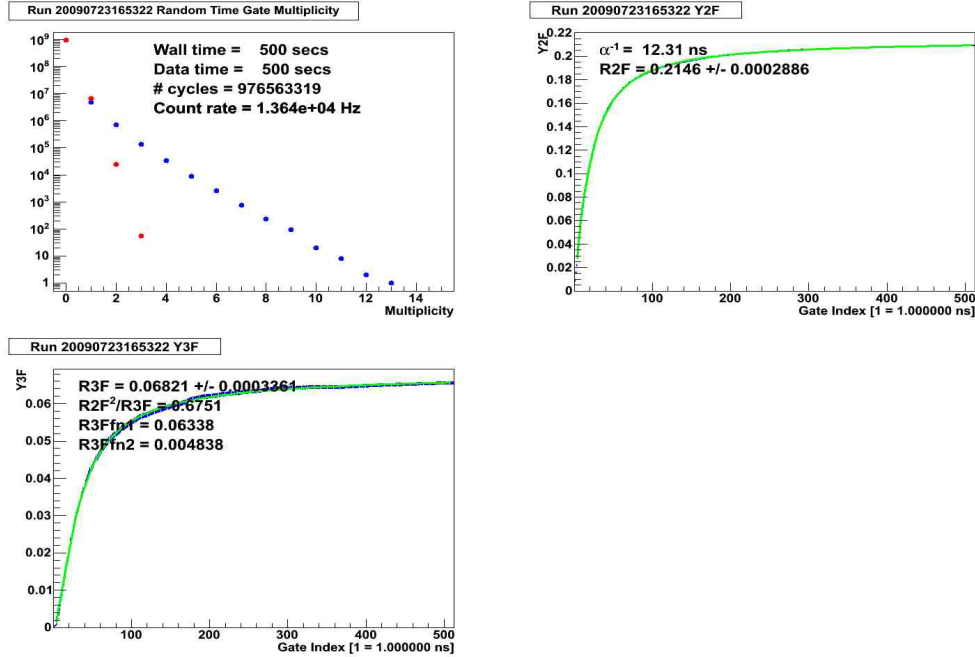
**Figure 2: 77 liquid scintillator detectors closely clustered around a source. Eight modules of two-by-four stand on the platform, one module of 13 is above the source on a low-mass stand. Distance from center of each detector to center of source is approximately 30 centimeters.**

We have been clustering up to 96 of these modules (usually 77) into a high efficiency configuration (Figure 2) estimated at 5.5% total efficiency for fast neutrons -- geometric efficiency of about 50% ("two-pi") times intrinsic detector

efficiency of a little more than 10%, after pulse shape discrimination. We digitize the pulses using Strück SIS 3320 8-channel 12-bit ADC boards at 6.25 nanoseconds per sample (160 mega-samples per second). Since each channel has its own buffer, the array is fully live as long as the rate is such that the individual channel pile-up rate is low. Based on the EJ-301 longest decay time of less than 1 microsecond, the single-detector pileup rate is less than 1% when the rate of all particles in the detector is less than about 10 kHz, so depending on the number of modules in our configuration, we can run at nearly one MHz with low pileup. Coincidentally, though the USB speed between the DAQ and computer turns out to be the current bottleneck, it allows us to keep up at almost one MHz when double-buffering on the ADC boards.

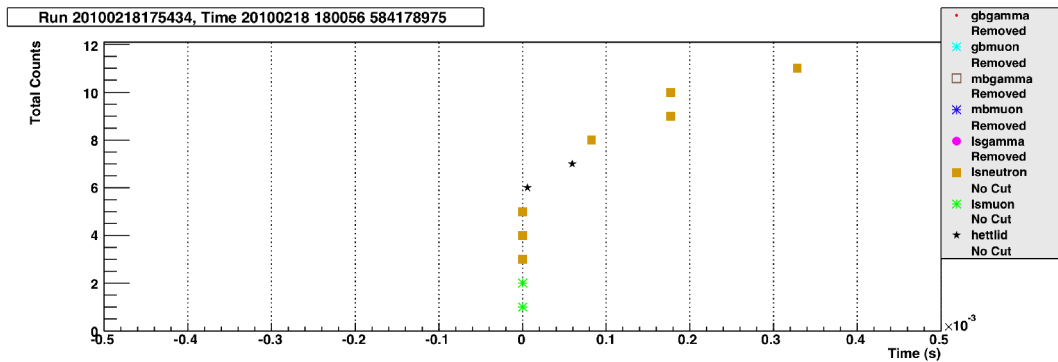
## Data Analysis and Source Characterization

Our method of pulse-shape discrimination currently relies on the ratio of tail to peak integrated counts as a “neutron score”. We bin events by gamma-equivalent energy and perform a two-gaussian fit to the neutron-score distribution. Then in each bin we draw a box so as to avoid the upper tail of the gammas’ distribution at a rate of 5-sigma. We also draw a 3-sigma lower tail on the neutrons’ distribution. Using this method, the fraction of “neutrons” that are actually misidentified gammas depends on the relative numbers of gammas and neutrons. With the 5-sigma gamma cut, the ratio of gammas misidentified as neutrons to real neutrons is about one-millionth of the ratio of gammas to neutrons in the bin.



**Figure 3:** (Upper left) fast neutrons accumulated in fixed gates of 512 nanoseconds, blue is data, red shows the expected Poisson (uncorrelated) distribution in 512 nanoseconds for the same rate. (Upper right) time-evolution of second Feynman moment, blue is data, green is fit. (Lower left) time-evolution of third Feynman moment, blue is data, green is fit.

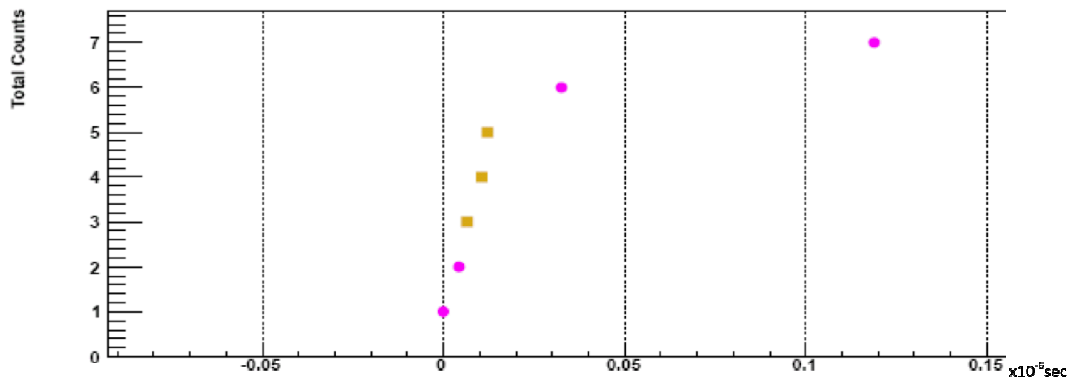
Figure 3 shows count distributions of fast neutrons to determine the mass of a plutonium ball. We analyzed the Feynman moments of correlated pairs and triples. The mass of a 2.4 kilogram plutonium ball determined from this data is 2.06 kg. The time dependence measured by the time gate dependence of the second moment is 12 nanoseconds. This is in extreme contrast to helium-3 data for the same bare Pu ball, which is 40 microseconds, characteristic of the need to moderate the neutrons before thermal capture. Because of the short time gates for fast counting, the total count time of 8-1/3 minutes gave almost  $10^9$  random time gate cycles, about  $10^3$  times more cycles than usual helium-3 counting, therefore much better statistics for the same count time. The analysis includes empirical corrections for single neutrons that make more than one count due to scattering between separate scintillator cells. These corrections are based on measurements of excess correlation using both Feynman moments and the distribution of intervals between events for the uncorrelated neutrons from an AmBe source and for simulations of our array. In our close configuration, the rate of double detected neutrons is about 1.7% for AmBe energy distribution, which contains higher energy neutrons than a typical fission source, and a much smaller triple-detection rate of 0.014%. This is close to the rate obtained from simulation of 1.5% for AmBe. The simulations for Pu showed a double-scattering rate of 0.55% from fission spectrum neutrons. We plan to obtain data with a lower-energy neutron source, a DD generator, later this summer.



**Figure 4: Muon-triggered fission chain restart burst in highly enriched uranium inside polyethylene and lead. The vertical axis shows accumulated counts since the first in the burst. Horizontal axis is elapsed time in milliseconds. Fast neutron counts are gold squares, thermal neutrons are black stars, and muons are green stars. The time across the plot is 1 millisecond, with basic time unit of 20 microseconds. On this time scale there are multiple restart fission chain fast neutron counts, spaced about 100 microseconds apart. Only one fast neutron is counted from two of them, and two fast neutrons from the middle one. The burst is spread over about 330 microseconds.**

Figure 4 shows an example of several particles detected from a single cosmic-induced fission event to show that we are seeing the time-evolution from a cosmic event, from a single fission, from a chain of fission events, and from restarts. Restarts occur when a fast neutron emerges from the fissile material (highly-

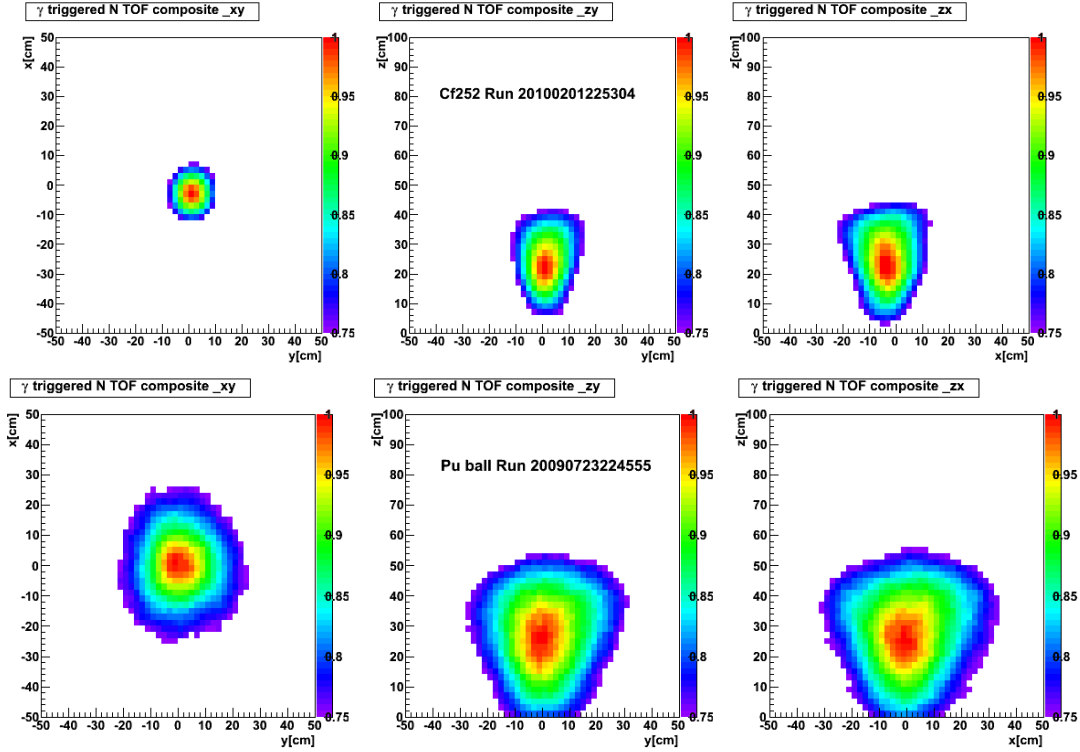
enriched uranium in this case) into a moderator, gets thermalized, and re-enters the fissioning material to induce a new fission chain.



**Figure 5: A single fission event in one shake (10 nanoseconds), followed by two gamma rays over 120 nanoseconds elapsed time.**

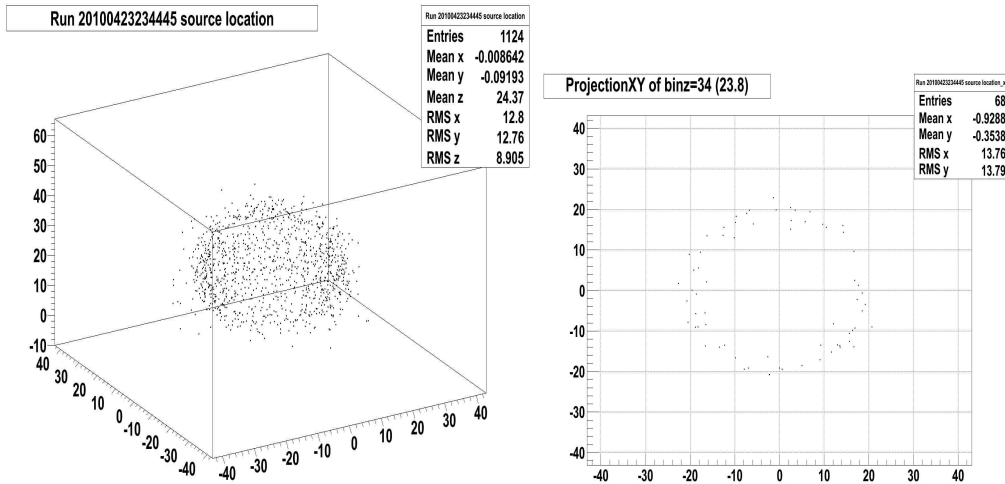
Even a burst of a few time-correlated particles is a good indicator of fission. Figure 5 shows a single fission event of gammas and fast neutrons taking place in 10 nanoseconds. We have made headway using these correlated gamma-neutron events to do time-of-flight imaging. Although imaging is an important capability outright, it is strong confirmation that we can detect individual fission events, because the method triggers on the most-immediate neutron after a gamma in any module, in other words, two different particles from the same fission. The gamma moves 30 cm/ns instantaneously compared to the fastest neutrons, 4.4 cm/ns of a 10 MeV neutron, the high-energy tail from fission. The neutron's module's location and estimated energy, along with the gamma's module's location and the delay between the gamma and neutron, define a hyperbolic surface. This surface is convolved with the probability distribution of the neutron's actual distance depending on its actual energy. The distribution of the source of the correlated events can be "built up" in time. Results with real data, but using the distance computed only from the deposited energy rather than a probability distribution, are shown in Figure 6.





**Figure 6: Projections in three dimensions of a real californium point source in 300 seconds, b) real data from a 3.5-cm diameter plutonium ball in 10 seconds. Note that the 3.5 cm ball is clearly larger than a point source.**

By using cuts of the most immediate neutrons in each detector following a gamma in any detector, we also have mapped out the surface of the emitter (Figure 7). We are also investigating other methods, including all neutrons and gamma-rays from individual spontaneous fission events.



**Figure 7: Surface and a cut through the surface of a simulated thin-shell spontaneous fissioning item, reconstructed from neutron-gamma correlation events.**

## **Plans**

We will spend the latter part of this year implementing the above-described theory into code. We have developed the theory for fast fission chain time evolution coupled with a moderation and diffusion model. We plan to develop an analysis code based on this fission chain theory that generalizes the current thermal neutron analysis to the fast counting regime. Further work also involves improving our estimates of all of the operating parameters of the 77-element array, including energy dependence of efficiency, multiple scattering rate of fission neutrons and gamma-rays, and improvements to the throughput at highest rates. We are also comparing these 3-inch by 4-inch cells of EJ-301 with EJ-309 in 3-by-4-inch, 6-by-6-inch, and 8-by-8-inch cells. The different cell sizes and different scintillators will require optimization of the neutron-gamma discrimination, and that exercise will improve our gamma-rejection with this array.

## **Acknowledgement**

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## **References**

1. V.V. Verbinski, W.R. Burrus, T.A. Love, W. Zobel, N.W. Hill, and R. Textor, 1968, Nuclear Instruments and Methods, 65, 8.